Rapid Communications

Microscopic dynamics of electron hopping in a semiconductor quantum well probed by spin-dependent photon echoes

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(Received 6 June 2019; published 4 September 2019)

Spin-dependent photon echoes in combination with pump-probe Kerr rotation are used to study the microscopic electron spin transport in a CdTe/(Cd,Mg)Te quantum well in the hopping regime. We demonstrate that, independent of the particular spin relaxation mechanism, the hopping of resident electrons leads to a shortening of the photon echo decay time, while the transverse spin relaxation time evaluated from pump-probe transients increases due to motional narrowing of spin dynamics in the fluctuating effective magnetic field of the lattice nuclei.

DOI: 10.1103/PhysRevB.100.121401

Understanding the charge and spin dynamics in condensed matter is essential for the development of novel spintronic devices in which the combination of charge transport with ultrafast spin initialization using optical pulses can be exploited [1–5]. In semiconductors, conduction band electrons which are localized on donor atoms or potential fluctuations, demonstrate long spin relaxation times due to the suppression of spin-orbit effects [6]. On the other hand, the hopping of electrons between localization sites or spin transfer between electrons due to an exchange interaction may become relevant and govern the spin dynamics [7]. The importance of the hopping of conduction band electrons was manifested in studies on the optical orientation, spin noise, and spatial diffusion of spin gratings where it was used to uncover the spin relaxation mechanism of electrons [8-10]. Transport effects such as the spin Hall effect in the hopping conductivity regime may occur in disordered two-dimensional systems which recently have attracted significant attention [11-13]. However, previous studies focused on the macroscopic properties of large ensembles, preventing insight into the local dynamics on the sub- μ m scale. Access to microscopic spin and charge dynamics has remained challenging also because the downscaling of an experiment down to the level of single carrier spins diminishes the correlations between quasiparticles. Therefore, the development of new approaches for the investigation of the microscopic charge and spin properties in large ensembles is in high demand.

Time-resolved optical techniques allow one to access the spin dynamics of both photoexcited and resident carriers [1,14]. Using them, one typically detects the macroscopic polarization of an ensemble of spins with a nonzero ensemble average, which was induced by a circularly polarized laser pulse due to the optical orientation of excitonic complexes. The most prominent examples for such techniques are polarized

photoluminescence [15,16] and pump-probe Faraday/Kerr rotation [17–20]. Recently, a novel technique based on spin-dependent photon echoes was introduced which potentially is well suited to investigate the spin dynamics of resident carriers in semiconductors [21–23]. The unique feature of photon echoes is the reversal of dephasing processes in an ensemble of emitters with an inhomogeneous broadening of optical transitions [24]. Thereby, echo techniques provide access not only to the homogeneous linewidth of the optical transition, but also show an exceptionally high sensitivity to spectral diffusion, e.g., due to an energy relaxation or resonance frequency variation [25]. However, this approach has not yet been applied to resident carriers so far.

In this Rapid Communication we demonstrate that by using spin-dependent photon echoes in combination with pumpprobe Faraday/Kerr rotation we can monitor the local spin dynamics of resident electrons and measure the hopping rate between the localization sites which so far had remained a free parameter in model descriptions. We evaluate hopping times in the order of several to tens of ns at low temperatures for electrons with a low density of $10^{10} \,\mathrm{cm}^{-2}$ in a CdTe/(Cd,Mg)Te quantum well (OW). If hopping is suppressed, both techniques give the same decay time of the coherent signal, which corresponds to the transverse electron spin relaxation. When the hopping rate, on the other hand, becomes comparable to the spin relaxation rate, the photon echo decay is accelerated. By contrast, in pump-probe, the decay time increases due to motional narrowing in the fluctuating effective nuclear magnetic field, enhancing the spin coherence. In full accord with the developed theoretical model, the hopping rate increases with increasing temperature while it decreases in the limit of stronger localization, e.g., when electrons are bound to donors as compared to electrons localized on potential fluctuations.

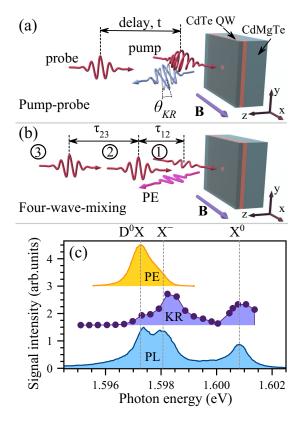


FIG. 1. Schemes of (a) pump-probe and (b) four-wave-mixing experiments including sketch of the investigated CdTe/(Cd,Mg)Te QW structure. The angle of Kerr rotation θ_{KR} or ellipticity is used to detect $S_z(t)$. (c) Spectral dependence of photoluminescence intensity and amplitude of long-lived signals measured using Kerr rotation and photon echo. Magnetic field B=0.25 T, temperature T=1.5 K.

The studied sample and the scheme of experiments are shown in Figs. 1(a) and 1(b). The semiconductor structure comprises a 20-nm-thick CdTe single QW sandwiched between Cd_{0.76}Mg_{0.24}Te barriers grown on a (100)-oriented GaAs substrate by molecular beam epitaxy. The unavoidable background of impurities results in low-density resident electrons that are localized both on potential fluctuations and on donors in the QW [23]. This is confirmed by the photoluminescence (PL) spectrum shown in Fig. 1(c), which shows three optical transitions that are attributed to the neutral exciton (X^0) centered at a photon energy 1.6008 eV, the negatively charged excitons (trion X^-) at 1.5981 eV, and the donor-bound excitons (D^0X) at 1.5972 eV, in accord with Refs. [23,26]. The sample was mounted in the variable temperature insert of a magneto-optical cryostat with the superconducting coil oriented for the Voigt geometry with the magnetic field B normal to the optical axis and parallel to the sample plane $(\mathbf{B} \parallel \mathbf{x})$. We studied the spin dynamics of resident electrons using time-resolved Kerr rotation (KR) and transient four-wave mixing (FWM). In both experiments resonant excitation of the exciton complexes was obtained with a tunable self-mode-locked Ti:sapphire laser as the source of optical pulses with a spectral width of 0.9 meV and duration of 2–3 ps at a repetition rate of 75.75 MHz. The optical pulses applied in each scheme have all the same central photon energy $\hbar\omega$ (degenerate configuration) and hit the sample close

to normal incidence with wave vectors \mathbf{k}_i , where i=1,2,3 is the pulse number in the sequences in Figs. 1(a) and 1(b). The excitation spot diameter was about 200 μ m and the pulse powers were kept low enough to remain in the linear regime with respect to single pulse excitation. Further details on the KR and FWM experimental setups can be found in Refs. [27,28].

In the KR experiment a circularly polarized pump pulse creates a macroscopic spin polarization of resident electrons along the z axis, with z being the sample normal [19,20]. In the external magnetic field the electron spins precess in the yz plane with the Larmor precession frequency $\Omega_L = g_e \mu_B B/\hbar$, where g_e is the electron g factor, μ_B is the Bohr magneton, and \hbar is the reduced Planck constant. The z component of the ensemble averaged spin density S_z is detected by the spin-Kerr and ellipticity effects, which result in corresponding variations of the polarization of the reflected, linearly polarized probe beam [see Fig. 1(a)]. Scanning the delay time t between the pump and the probe allows us to measure the spin dynamics of the macroscopic spin polarization and to determine both the frequency Ω_L and decay time $T_{2,\mathrm{KR}}^*$ by fitting the experimental data with $S_z(t) \propto \cos{(\Omega_L t)} \exp{(-t/T_{2,\mathrm{KR}}^*)}$.

In the transient FWM experiment a sequence of three laser pulses is used to generate a coherent optical response along the phase-matching direction $\mathbf{k}_{\text{FWM}} = \mathbf{k}_3 + \mathbf{k}_2 - \mathbf{k}_1$, where in our case $\mathbf{k}_2 = \mathbf{k}_3$ [29]. Due to significant inhomogeneous broadening of the optical transitions this response is given by photon echoes (PEs). Here, we focus on the three-pulse PE, which appears at a delay time $t_{PE} = 2\tau_{12} +$ τ_{23} relative to the arrival time of pulse 1, where τ_{ij} is the delay time between pulses i and j. For resonant excitation of X^- or D^0X complexes [see Fig. 1(c)], PEs appear even for long delays, decaying on the timescale of several ns when τ_{23} is scanned [Fig. 2(a)]. This decay time is significantly longer than the lifetime of the optical excitations below 100 ps [23]. Here, the pulse 1-2 sequence orients the spins of each resident electron depending on the excitonic resonance frequency ω_0 and the delay time τ_{12} . As a result, a spin grating in coordinate and frequency space is formed,

$$S_y^0 - iS_z^0 \propto i \exp\left(\frac{i\Omega_L \tau_{12}}{2}\right) \cos\left(\omega_0 \tau_{12} + \mathbf{k}_{2,\parallel} \mathbf{r} - \mathbf{k}_{1,\parallel} \mathbf{r}\right), \quad (1)$$

for orthogonally linearly polarized pulses 1 and 2 with the in-plane components of the wave vectors $\mathbf{k}_{1,\parallel}$ and $\mathbf{k}_{2,\parallel}$, respectively. The spin grating is retrieved optically with the third pulse, which induces the three-pulse PE [22]. The time evolution of the grating distribution (1) before the arrival of pulse 3 is characterized by the electron Larmor precession and the decay of the spin grating proportional to $\cos(\Omega_L t) \exp(-\tau_{23}/T_{2,\text{PE}}^*)$. In contrast to the established transient spin grating technique [3,10], where $\tau_{12} \approx 0$, the spin-dependent PE appears when $\Delta\omega_0\tau_{12}\gg\pi$ with $\Delta\omega_0$ being the spectral width of the addressed optical transitions. Therefore, the PE signal is very sensitive to spectral diffusion of the resident electrons, which is important in the hopping regime. Variation of the delay time τ_{23} allows one to extract Ω_L and $T_{2,PE}^*$ using the above form for fitting, similar to KR. Note that we measure the absolute value of the electric field amplitude at the PE peak maximum and, therefore, the signal

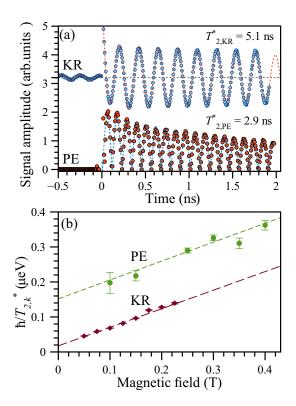


FIG. 2. (a) Examples of pump-probe Kerr ellipticity (labeled KR) and PE transients measured for resonant trion (X^-) excitation at 1.5981 eV. T=1.5 K and B=0.2 T. The three-pulse photon echo amplitude is measured for a fixed delay $\tau_{12}=26.7$ ps. Dashed curves show fits using an exponentially damped oscillatory function with $\Omega_L=28.1$ rad/ns for both traces, as well as $T_{2,\mathrm{KR}}^*=5.1$ ns, and $T_{2,\mathrm{PE}}^*=2.9$ ns. (b) Magnetic field dependence of relaxation rate for X^- at 1.5981 eV obtained from the KR and PE signals. T=1.5 K, $\tau_{12}=26.7$ ps. Results are fitted using a linear field dependence (dashed lines) with the same $\Delta g_e=9\times 10^{-3}$, as well as $T_{2,\mathrm{KR}}\gtrsim 20$ ns, and $T_{2,\mathrm{PE}}=4.3\pm0.7$ ns.

is described by the modulus of the oscillatory function, as shown in Fig. 2(a) [28].

The spectral variation of the signal amplitudes of the resident electron relaxation dynamics is shown in Fig. 1(c). In contrast to PE, where long-lived signals are observed only for resonant excitation of X^- and D^0X , a long-lived KR signal is present also for the resonant excitation of neutral excitons, because even in a degenerate pump-probe experiment the excitation and detection of spin polarization may be performed at different optical transitions. First, we concentrate on the resonant excitation of trions, where the differences in relaxation times between the KR and PE results are most pronounced. The KR and PE transients are shown in Fig. 2(a). Both signals contain additional contributions from the spin dynamics of optically excited carriers with short response times below 100 ps. Here, we concentrate on the long-lived spin dynamics, which is contributed only by resident electrons. One expects to observe the same relaxation behavior in KR and PE, as the decay is governed by spin dephasing of the electron ensemble, i.e., $T_{2,PE}^* = T_{2,KR}^*$. Indeed, the Larmor precession frequencies at B = 0.2 T are the same, $\Omega_L = 28.1$ rad/ns, in both cases. However, the decay times are surprisingly different. For KR

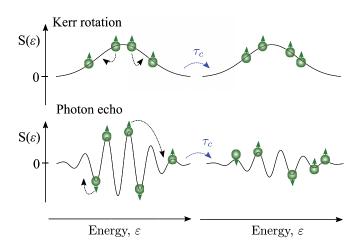


FIG. 3. Scheme of energy distribution of spin density $S(\varepsilon)$ of localized resident electrons in the KR (top) and PE (bottom) experiments before (left) and after (right) hopping between localization sites. A single hop is accompanied by a change of energy ε , while spin is conserved. Therefore, hopping destroys the spectral grating, but does not influence the macroscopic spin.

we obtain a notably longer time $T_{2,\mathrm{KR}}^*=5.1$ ns as compared to the shorter PE decay time $T_{2,\mathrm{PE}}^*=2.9$ ns. Spin dephasing in a transverse magnetic field can result from the spread of electron g factor Δg_e [27]. To account for this contribution, we measured the KR and PE signals for different magnetic fields and evaluated the dependences of Ω_L and dephasing times on B. Both methods give identical B-linear dependencies of Ω_L , from which we obtain $|g_e|=1.60$, in agreement with previous reports [23,27,30,31]. Details are given in the Supplemental Material [32].

The magnetic field dependencies of the decay rates $1/T_{2,\mathrm{PE}}^*$ and $1/T_{2,\mathrm{KR}}^*$ are plotted in Fig. 2(b). Both show a linear rise with increasing magnetic field and can be described by $1/T_{2,k}^* = 1/T_{2,k} + \Delta g_e \mu_B B/\hbar$ (k is KR or PE), with the same slope corresponding to $\Delta g_e = 9 \times 10^{-3}$. This confirms that the KR and PE signals are provided by the same subensembles of resident electrons. However, the relaxation times $T_{2,\mathrm{KR}} \gtrsim 20$ ns and $T_{2,\mathrm{PE}} \approx 4$ ns are drastically different.

The drastic difference of the PE and KR decay times is related to the specific impacts of the localized electron dynamics on the coherent response, as sketched in Fig. 3. The hopping of electrons between localization sites destroys the spin grating in Eq. (1): Once an electron leaves its initial site and arrives at a site with different location \mathbf{r} and frequency ω_0 , it no longer contributes to the PE signal. However, it continues to contribute to the KR signal provided that spin is conserved during the course of hopping, since hopping does not change the macroscopic spin polarization and the variation of energy $\Delta E_{\text{hop}} \sim k_B T$ is small compared to the spectral width $\Delta \omega_0$. We emphasize that the electron displacement by hopping is small and therefore the spectral diffusion plays a decisive role in the decay of PE signal. In our OW the electron spin coherence is controlled by the hyperfine coupling with nuclear spins [6,33]. If the hopping processes, characterized by the electron spin correlation time at the localization site τ_c , are efficient such that the electron spin precession rate in the

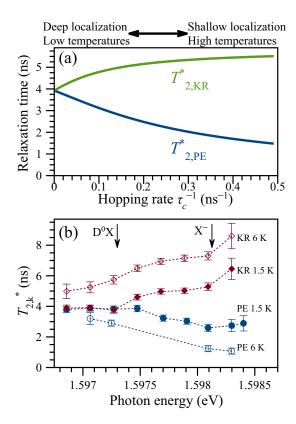


FIG. 4. (a) Calculated dependence of relaxation times $T_{2,k}^*$ on hopping rate τ_c^{-1} for $\delta_e=0.1~{\rm ns}^{-1}$, $\Delta g_e=9\times 10^{-3}$, and $B=0.2~{\rm T}$. (b) Spectral dependence of relaxation times $T_{2,k}^*$ measured at T=1.5 and 6 K for $B=0.2~{\rm T}$.

field of nuclear fluctuations $\sqrt{\langle \Omega_N^2 \rangle}$ is small, $\sqrt{\langle \Omega_N^2 \rangle} \tau_c \lesssim 1$, macroscopic spin relaxation in the ensemble takes place after several hops only with a rate given by $T_{2,\mathrm{KR}}^{-1} \sim \langle \Omega_N^2 \rangle \tau_c$ [34,35] due to the effect of motional narrowing. By contrast, the PE decays with the time constant $\sim \tau_c$ independent of the spin relaxation mechanism, resulting in $T_{2,\mathrm{PE}} \sim \tau_c \ll T_{2,\mathrm{KR}}$, in agreement with our findings. The microscopic theory based on the kinetic equation for the spin distribution function in the Supplemental Material [32] (see also Refs. [35–41] therein) gives the following expressions for the decay times,

$$T_{2,\text{KR}} = \frac{2 \exp\left[-1/(\delta_e \tau_c)^2\right]}{\sqrt{\pi} \delta_e \operatorname{erfc}\left[1/(\delta_e \tau_c)\right]}, \quad T_{2,\text{PE}} = T_{2,\text{KR}} - \frac{2}{\delta_e^2 \tau_c}, \quad (2)$$

with $\delta_e^2 = 2\langle \Omega_N^2 \rangle / 3$ and erfc (x) being the error function. The results of numerical calculations are shown in Fig. 4(a), where the relaxation times $T_{2,k}^*$ are plotted as a function of the hopping rate τ_c^{-1} .

The strength of localization governs the hopping rate of the resident electrons. Therefore, it is possible to address different hopping regimes by optical excitation of D^0X and X^- , i.e., electrons bound to donors (stronger localization) or electrons localized on potential fluctuations (weaker localization), respectively. Furthermore, an increase of temperature will lead to an increase of the hopping rate. Consequently, the difference between the relaxation times $T_{2.KR}$

and $T_{2,PE}$ should become even more pronounced. In order to test this conjecture, we have studied the spectral and temperature dependences of $T_{2,KR}^*$ and $T_{2,PE}^*$ at B=0.2 T [Fig. 4(b)].

First, we observe that for resonant excitation of D^0X (1.5972 eV) at T = 1.5 K the relaxation times $T_{2.\text{KR}}^*$ and $T_{2,\text{PE}}^*$ are identical with a value of 4 ns. This is due to the stronger localization of donor bound electrons and therefore the regime of $\langle \Omega_N^2 \rangle \tau_c^2 \gg 1$ is realized in this case [8,32,33]. Here, the electron spin is lost efficiently at a given donor via the hyperfine coupling and, consequently, hopping is unimportant so that $T_{2,PE}^* = T_{2,KR}^*$. Excitation with a larger photon energy addresses electrons with a weaker localization and consequently $T_{2,KR}^*$ increases while $T_{2,PE}^*$ decreases, in accordance with our predictions. Second, a temperature increase leads to a similar behavior, which also excludes a possible origin of the τ_c behavior in the exchange interaction between the resident electrons, because it should be largely independent of temperature. Using the theoretical results in Fig. 4(a) we determine the hopping time of electrons localized on potential fluctuations to be $\tau_c \approx 5$ and 2 ns for T = 1.5and 6 K, respectively. For electrons bound to donors we observe hopping only at T=6 K with $\tau_c \approx 5$ ns and $\delta_e \approx$ $0.1\,\mathrm{ns^{-1}}$. The magnitude of δ_e is in full accord with the estimate which is given in the Supplemental Material [32] and Refs. [6,42–44].

In conclusion, we have demonstrated that spin-dependent photon echoes represent a powerful tool to access directly the local spin and charge dynamics of resident carriers in semiconductors. Our findings allow one to determine the key transport parameter of a localized system, the hopping rate τ_c by purely optical means, thereby bridging the gap between optical and transport spectroscopy. The electron spin correlation time τ_c directly controls the dynamical nuclear polarization induced by the hyperfine interaction [6,15], opening up prospects to optimize nuclear spin memories by tailoring electron localization. Finally, we observe suppressed hopping of donor bound electrons at a low temperature of 1.5 K where the spin relaxation of the ensemble takes place in the fluctuating nuclear fields. This suggests that the decay of spin-dependent photon echoes from donor bound excitons can be further extended by several orders of magnitude using spin echoes, which is attractive for applications in quantum communication.

We are grateful to O. S. Ken and V. L. Korenev for useful discussions. We acknowledge the financial support by the Deutsche Forschungsgemeinschaft through the International Collaborative Research Centre TRR 160 (Project A3). S.V.P. thanks the Russian Foundation for Basic Research (RFBR) (Project No. 19-52-12046) and St. Petersburg State University (Project No. 11.34.2.2012 and Grant No. 40847559). L.E.G. and M.M.G. acknowledge partial support from RFBR (Project No. 19-52-12038); L.E.G. is grateful to the Foundation for the Advancement of Theoretical Physics and Mathematics "BASIS." The research in Poland was partially supported by the Foundation for Polish Science through the IRA Programme cofinanced by the EU within SG OP and by the National Science Centre through Grants No. 2017/25/B/ST3/02966 and No. 2018/30/M/ST3/00276.

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